A PROPOSED RAPID METHOD FOR MEASURING AREA METHANE EMISSIONS: AN EXPLORATORY APPLICATION IN MANHATTAN, NEW YORK, USA

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Abstract

Methane is an important greenhouse gas, but methane emissions are poorly understood, in large part due to limited atmospheric methane data on local scales. Local and regional scale methane emissions data are urgently needed to improve modeling of future climate change, and support energy plans and policies to minimize future climate impacts of socio-economically needed energy utilization. There have been numerous recent reports on local ground-level ambient air methane surveys that have provided more thorough data on methane sources in some urban areas. Such surveys generate substantial amounts of high quality ground-level methane concentration data, usually with reliable time and geo-referenced location data. We examined the potential usefulness of such data sets for generation of estimates of methane emissions for surveyed areas. Our efforts focused on development of a generally applicable, relatively simple mass-balance approach to estimate area methane emissions from mobile, ground level ambient air methane concentration and local weather data. The data examined were collected in Manhattan, New York, USA over 5 days in late 2012. Using the ratio of methane emissions (μg $m^{-2}s^{-1}$) to natural gas usage (μg $m^{-2}s^{-1}$), the resulting methane emissions estimates for Manhattan were compared to 5 other cities (emissions reported by other investigators using other methods). The emissions estimates for Manhattan derived from ground-level mobile methane surveys were within the range of the estimates for the other cities. In addition, the emissions rates reported for the cities indicate natural gas should not be considered more climate-beneficial than other fossil fuels.

Keywords: Methane emissions, New York

Introduction

Increasing levels of greenhouse gases in the atmosphere are expected to cause the global climate to warm substantially, with associated impacts on human health, the environment and economic systems (Stocker et al., 2013). Methane is a major, potent greenhouse gas (GHG) and the predominant (>85 vol%) hydrocarbon in natural gas (NG). Unlike emissions of carbon dioxide, regarded as the predominant greenhouse gas, methane emissions cannot be reliably estimated from fossil fuel consumption data. Rapidly increasing development of unconventional NG resources, and the state of repair of NG infrastructure have raised increasingly urgent concerns that methane emissions, and associated climate impacts, are being substantially underestimated. Studies have found leakage from commercial NG system infrastructure is a substantial source of methane emissions, but the amount of data available is limited (Zimnoch, Godlowska, Necki & Rozanski, 2010; Alvarez, Pacala, Windbrake, Chameides & Hamburg, 2014; Jackson et al., 2014; Phillips et al., 2012). Methane emissions estimates based on technologically up-to-date measurements of actual methane levels in the atmosphere at the local to regional scale are urgently needed (Bellucci, Bogner & Sturchio, 2012; Dlugokencky, Nisbet, Fisher & Lowry, 2011; Fowler,1999; Gan et al., 2010;The White House, 2014; Howarth, 2014; Alvarez, 2012; EPA-OIG, 2013).

Mobile Cavity Ring-Down Laser Spectrometers (CRDS) mounted in motor vehicles provide rapid, convenient and accurate methane measurements. Recent mobile CRDS investigations have focused on leak detection, finding on average a gas leak every 400 meters of city streets (Phillips et al., 2012; Jackson et al, 2014). Such leak surveys generate large amounts of accurate, time- and location- referenced, methane concentration data. Estimates of methane emissions based on actual, technologically up-to-date measurements, such as those provided by mobile CRDS, could help fulfill the urgent need for such estimates to enable more informed policies and effective management of NG (Bellucci et al., 2012; Dlugokencky et al., 2011; Fowler,1999; Gan et al., 2010; The White House, 2014; Howarth, 2014; Alvarez, 2012; EPA-OIG, 2013). To our knowledge, how mobile CRDS survey data might be used to estimate area methane emissions in urban settings has not been examined or reported.

We explored the application of a simple mass-balance approach to estimate methane emissions from mobile CRDS ground level ambient air methane concentration and local weather data. It was our intention to maintain the greatest practical transparency regarding the approach, data analysis, results and conclusions. Consequently, we intentionally avoided complex or less transparent statistical analytical methods. To our knowledge, this is the first published methane emissions method based on ground level mobile CRDS methane concentration data. The method is rapid and does not require use of aircraft, or long term, static installations of expensive equipment. Hence, we anticipate fuller development of this method could facilitate rapid collection of the methane emissions data urgently needed to improve current climate change modeling, NG utilization and management, and climate oriented energy plans and policies.

Materials and methods

Conceptual Approach

In order to determine the methane emissions for a selected area over a given time it would be necessary to sum all the emissions from all point and nonpoint sources, an impractical challenge for all but the smallest areas. Alternatively, it should suffice to calculate the flow of methane carried in the air moving over the area of interest (Fowler, 1999). In urbanized areas advection by wind and turbulence within the planetary boundary layer (PBL) can be reasonably expected to assure relatively rapid and thorough mixing from the surface to the top of the PBL (Gan et al., 2010). Calculation of the net bulk flow of methane does not require detailed micrometeorological data or data on the internal dynamics of the PBL. Assuming (i) wind advection is the major determinant of horizontal transfer, (ii) turbulence in the PBL the main determinant of vertical transfer, and (iii) vertical transfer ends at the top of the PBL, enables selection of a 3-dimensional space through which all gas emissions from the underlying land surface must move.

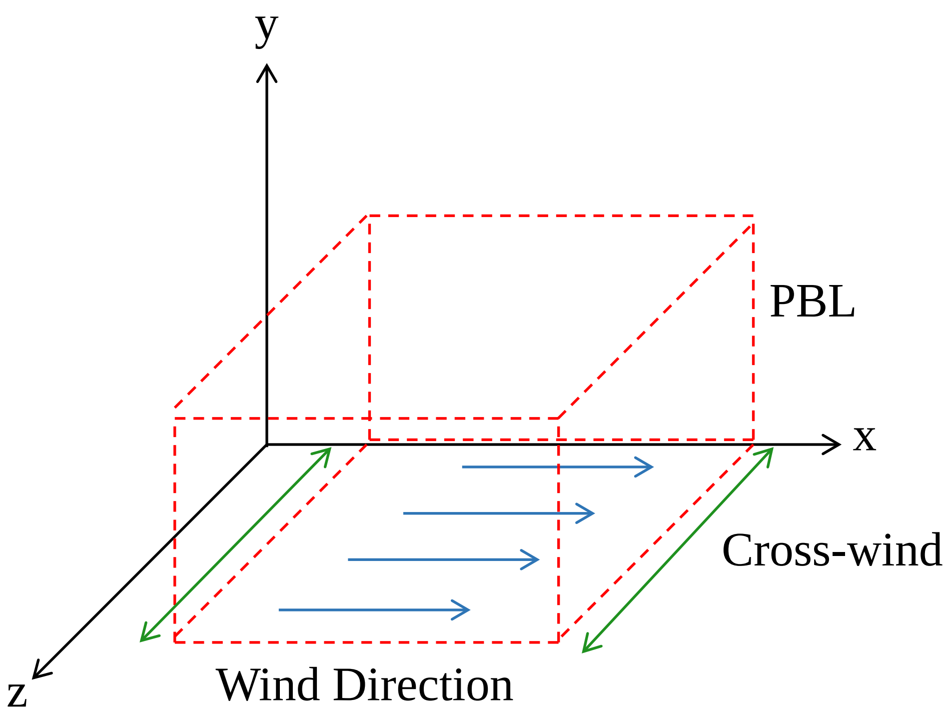

Figure 1. Main components of simple, three dimensional, mass balance box overlying rectangular surface(x-z plane) with methane emissions sources (sinks).

For simplicity of discussion we consider a simple rectangular land area as the emissions source area and ground-level methane data available from survey runs along the upwind and downwind sides of the source area (Figure 1). An overlying three-dimensional (x,y,z axes in Figure 1) "box" is designated with the height of the PBL (y), the width (x) equal to the wind-run distance between the upwind and downwind methane survey runs, and length (z) equal to the length of parallel cross-wind upwind and downwind methane survey runs. We assume negligible net wind or methane flow through the lateral walls or the top of the box (top of the PBL). The air flow rate through the upwind and downwind walls of the box are then necessarily equal as a mass balance requirement. The volume of air flow through the box can then be calculated from the crosswind length (z dimension) of the box, the PBL height (y dimension) and the wind speed (velocity along x-axis, x interval per unit time). The net methane emissions from (or removal by) the land area on the floor of the box is then the product of the volumetric rate of air flow through the downwind wall and the change in average methane concentrations between the upwind and downwind survey runs of the estimation area.

The approach requires or assumes the following:

1. Wind speed and direction data.
2. Negligible net flow of methane through the top of the PBL.
3. Sufficient methane concentration data to allow calculation of methane concentrations in well mixed air along the upwind and downwind sides of the target measurement area.
4. Wind and weather conditions that favor rapid horizontal and vertical dispersion of emitted methane throughout the PBL so that the methane concentration in well mixed ground level air will be effectively equal to the methane concentration along any overhead horizontal transect in the PBL.[1]

Generally, there is limited mixing between the PBL and overlying atmosphere (Piironen, 1996). Meteorological data, including PBL data, for Manhattan were available at http://nycmetnet.ccny.cuny.edu (Legbandt, T.) Our street survey of Manhattan provided ground level methane data.

Well and Poorly Mixed Air: Data Filtering

The reliability of a simple mass balance approach based on mobile platform CRDS ground level methane measurements depends on the assumption that the sampled air is well mixed. Initially we accepted the assumption that point sources of methane were sufficiently infrequent in overall methane survey data sets that they would have minimal impact on estimates based on a mass balance approach (Payne and Ackley, 2013). However, in the present effort we were working with a previously collected data set, originally designed only to scout for gas leaks, not to estimate methane emissions. Consequently, we had to select emissions estimate target areas for which small subsets of the methane survey data met the requirements for the mass balance approach. Each such area is only a small portion of the much larger methane survey data set. As the size of the emissions target area becomes smaller, the potential for overestimation errors due to high methane readings in poorly mixed air associated with point sources near the survey path increases. Consequently, it was necessary to find a means of filtering the available data to remove effects of sampling poorly mixed air.

For the purposes of a simple mass balance approach, well mixed air is that which is commonly present in the local sampling area without direct influence of nearby methane sources on methane concentration. The

[1] This condition is likely met in the PBL over Manhattan, but will often not be met in other environments. In those cases, it appears likely that there will be a decrease in methane concentration with increasing altitude in the PBL (Zimnoch, 2010; Mays, 2009). The present approach will remain useful in such other environments provided a reliable algorithm for change in methane concentration with altitude in the PBL is available or can be developed.

methane concentration in well mixed air will vary due to turbulence and other factors, but collectively the variations will be randomly distributed within a relatively narrow range around the well mixed air mean methane level. Methane emissions from sources sufficiently far upwind of the sample location will be well mixed into the air. Methane from nearby sources will be apparent as excursions from the typical range for local well mixed air. The time and space duration of the methane concentration excursion due to a local source depends on the emission rate from the source, local physical features, and wind and atmospheric conditions (Jackson et al., 2014). Similarly, excursions to methane levels below locally typical levels can occur due to turbulence related intrusions of air from outside the local vicinity (edge effects), or temporary excursions in instrument function. Our direct examinations of CRDS methane survey data from Manhattan and numerous other areas confirm these typical variation patterns and relationships for well mixed and poorly mixed air. Given the amounts of data involved, direct examination of all the variations in methane survey data sets is not practical. We developed a relatively simple method for distinguishing well mixed air from poorly mixed air data.

Our data filter is based on recognition that the variations in methane concentrations in well mixed air will be limited in range, relatively consistent, and generally symmetrical in the rise to and fall from maxima (or minima). Variations in poorly mixed air will be larger and the ascent to and descent from maxima will be asymmetrical. It follows that if a data set is comprised of values with consistent, symmetrical, random variations around a central value, the fractiles (or quantiles) of that data set should vary consistently and uniformly, which we regard as an aspect of quantile regression analysis (Koenker, R., & Hallock, K., 2001; Cade, B. S., & Noon, B. R., 2003). In the present methane data case, a plot of methane concentration by fractiles for well mixed air should be a straight line. If the range of the data set is the same as each of the fractiles, then such a plot will have no slope. If the range of the data set is greater than that of each fractile, and the approach to and descent from maxima is symmetrical, then the methane-fractile plot will be a straight line with positive slope. If data with asymmetrical approaches to maxima (and minima) occur in the data set they will cause the methane-fractile plot to deviate from linearity.

We found the use of 100-fractiles, i.e., percentiles, to be convenient in that it provides reasonable resolution while facilitating visual accessibility and data processing. Examination of numerous methane survey data sets using methane-percentile plots confirmed the expected linearity and deviations in every case. We used the Excel linear trend line capability (least squares best fit) iteratively to adjust the low and high percentile bounds on the trend line until r^2 was maximized. Linear regressions could always be bounded to have r^2 greater than 0.98, usually greater than 0.99 (Figure 2.). The equation of the maximized-r^2 trend line was then evaluated at the 50th percentile to determine the representative methane level for well mixed air along that methane survey path.

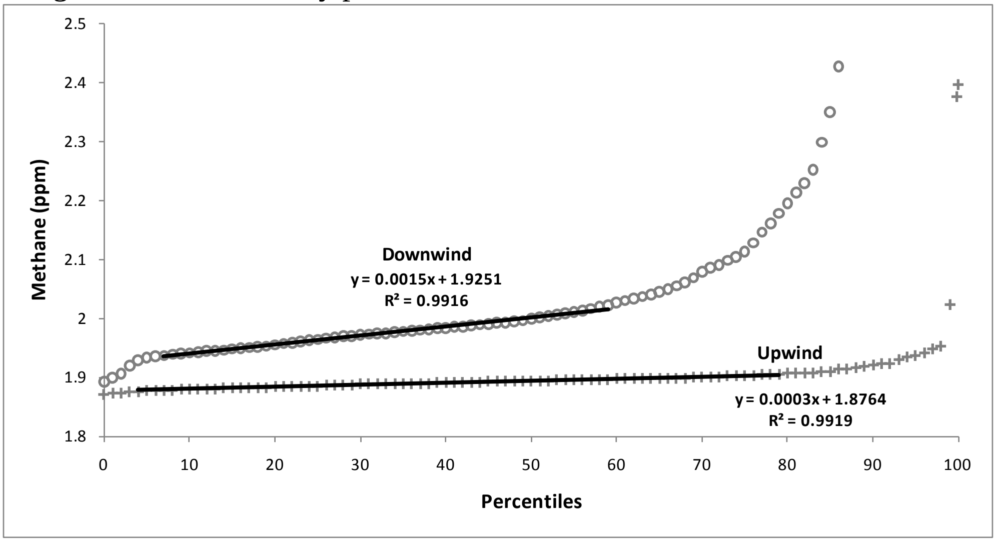

Figure 2. Methane-percentiles plots for Upwind and Downwind ground level methane survey data sets for the Southeast Lower Manhattan methane emissions estimate area (discussed below).

The mass balance approach requires comparison of methane levels at upwind and downwind locations. In practice the amount of data available for upwind and downwind locations is often not the same. The use of methane-percentile plots to examine every data set of interest effectively normalizes all the data sets to a basis with which most investigators are familiar, facilitating comparisons and communications.

Ambient Air Methane Measurements

Over a period of 5 days from 27-30 November and on 9 December 2012 we surveyed a total of 165 miles of streets in Manhattan, NYC, NY (Payne Jr., B.F. & Ackley, R., 2012, December 12) in the manner reported by Jackson et al. (2014) for their gas leaks survey of the streets of Washington, D.C. We used the same Cavity Ring-down Spectrometer (CRDS) and the same measurement and calibration procedure, run by the same operator (Robert Ackley). We have found under mobile field use conditions the CRDS instruments we have used to be reliably sensitive to ±2% relative and a typical minimally sized data set (N>40) for areas with stable methane levels to have a 99% (alpha = 0.01) confidence interval around 0.1% relative.

Methane Data Processing

The data were compiled and processed using Microsoft Excel and a program (which we refer to as XTL) in R developed by Xiaojing Tang of Boston University with subsequent revisions by Jacob Lysinger. The program reduced the amount of manual data processing required by performing five tasks. XTL reduced the size of the raw data sets by deleting instrument function parameters and reducing reported accuracies to appropriate levels, .e.g., 0.001 ppm for methane. XTL moved any data records that included out of range methane levels to a separate file. The normal range limits are adjustable and were set to collect data from reference gas runs to check instrument function or other unusual deviations in instrument behavior. XTL eliminated redundant data that accumulated during necessary stops of the survey vehicle due to traffic or other operational requirements. XTL used GPS data to identify methane data for any stop lasting more than 1 second, calculated the average methane concentration for that stop, moved all the original data for the stop to a separate file and inserted a single entry of the average methane concentration for that location back into the data set. XTL then determined the maximum, minimum, average, median and 1st to 99.9th percentiles for the processed data set. XTL always generates at least 3 files: a program processing log, the processed methane survey data set, and a statistical summary. XTL may generate more files if there are out of range data or stop location data. We elected to set the maximum size of output files at 100,000 lines. Consequently, for large raw data sets XTL will report the whole processed data file as a sequential series of 100,000-line files.

Mixing in the Planetary Boundary Layer and Meteorological Data

On 19-20 June 2013 we undertook a limited investigation of the lower portion of the PBL. We measured methane concentrations at ground level and outside 8 buildings in Manhattan at elevations from 20 to 164 meters above street level. Analysis of the data indicated methane concentrations were consistent from street level to a height of at least 164 meters above street level. Based on that data and ceilometer data we concluded that it was reasonable to treat the PBL (mixing layer) as uniformly mixed at the sampling times of concern.

The height of the PBL during relevant sampling periods was estimated from ORSL ceilometer data (retrieved from http://sky.ccny.cuny.edu/wc/Thales/index.php or processed and supplied directly by the ORSL).

Results and discussion

Winds and the Planetary Boundary Layer

Winds were generally northeasterly (NE, bearing 20°-70°, 3-day average 40°) on 27 and 30 November and 9 December, and generally west-southwesterly (WSW) on 28 November (bearing 270°-290°) and 29 November (230°-260°, 2-day average 240°). Average wind speeds were similar for the two wind-direction data sets (average 3.0 $m \cdot s^{-1}$ for NE wind days, and 3.4 $m \cdot s^{-1}$ WSW wind days), but those averages contained wide variations (1.3-5.8 $m \cdot s^{-1}$) and 35% of wind direction data values were missing or “variable”. Average wind speed during the sampling time for a given data run serves as the net horizontal movement proxy value in the mass balance methane emissions calculation. It was, therefore, necessary to better account for “variable” wind direction data. Wind data entries with “variable” direction data typically have no associated wind speed data. Average wind speed could be calculated using only wind speed/direction pairs that were not “variable”. However, a “variable” wind data entry implied wind was not blowing consistently in any predominant direction, and, hence, there was no net horizontal displacement of air during that time interval. Consequently, we set wind speed associated with any period of “variable” direction to zero and included that wind data in calculation of the average wind speed. This reduced the average wind speed in proportion to the number of “variable” wind direction entries in the wind data record for each emissions estimate area, better reflecting the impacts of periods with no net horizontal movement of the air and methane it contained.

During sampling on the two WSW wind days (28 and 29 November) the PBL was relatively stable with a height around 800 meters. During NE-winds sampling on 30 November the PBL was reasonably stable at a height of around 1000 meters, but over a period of 4 hours on 27 November the PBL collapsed to a height of around 100 meters apparently in association with a drop in barometric pressure (30.13-30.06 in.Hg, 1.007-1.004 atm).

Indications from the Methane Survey Data

Multiple analyses of the available methane survey data for the whole island indicated about half the methane emissions likely occurred south of latitude 40.755 (east-west line roughly through the end-to-end center of 42nd Street). Methane emissions appeared to be clustered in the lower East Side, and in areas where construction is densest, especially 34th Street to Central Park, and in the Financial District.

The methane survey was not designed to collect data for the purpose of estimating methane emissions; hence, most of the collected data was not appropriately positioned in space and time to support estimates of methane emission rates. We undertook detailed examination of the data files in an attempt to identify data subsets that contained reasonably time and space related methane survey data to generate more quantitative estimates of methane emissions for the areas covered by such subsets.

The simple mass balance approach we propose suggests the net methane emissions from (or removal by) the target land area is the product of the volumetric rate of air flow across the downwind edge of that land area and the difference between representative (linear trend 50th percentile) methane concentrations in well mixed air for survey runs along the downwind and upwind sides of the land area. Consequently, data sets are well positioned in time and space when they run along areas that are upwind and downwind with respect to each other, and both were collected within a reasonable time frame.

Ideally the time between the upwind and downwind runs would be the time required for the wind to traverse the area between. This ideal time separation is difficult to achieve under field conditions as timing of runs may be entirely determined by traffic, etc. In most urban areas the numbers, locations and emission rates of methane sources within a target measurement area are not likely to change over time intervals in the range of a few hours to days. However, wind and PBL dynamics determine where and how fast methane from emissions sources moves. Consequently, appropriate time intervals between up- and downwind runs, is often determined by the duration of consistent wind conditions and PBL height, either or both of which can change over periods as short as a few minutes. For this investigation of our proposed simple mass-balance method, we searched for data subsets comprised of runs with reasonable time and distance separations and reasonably consistent wind and PBL conditions.

Our search encountered the areas marked in Figures 3 and 4. The methane survey data for these areas was sufficient to support experimental application of our simple mass balance emissions estimate method.

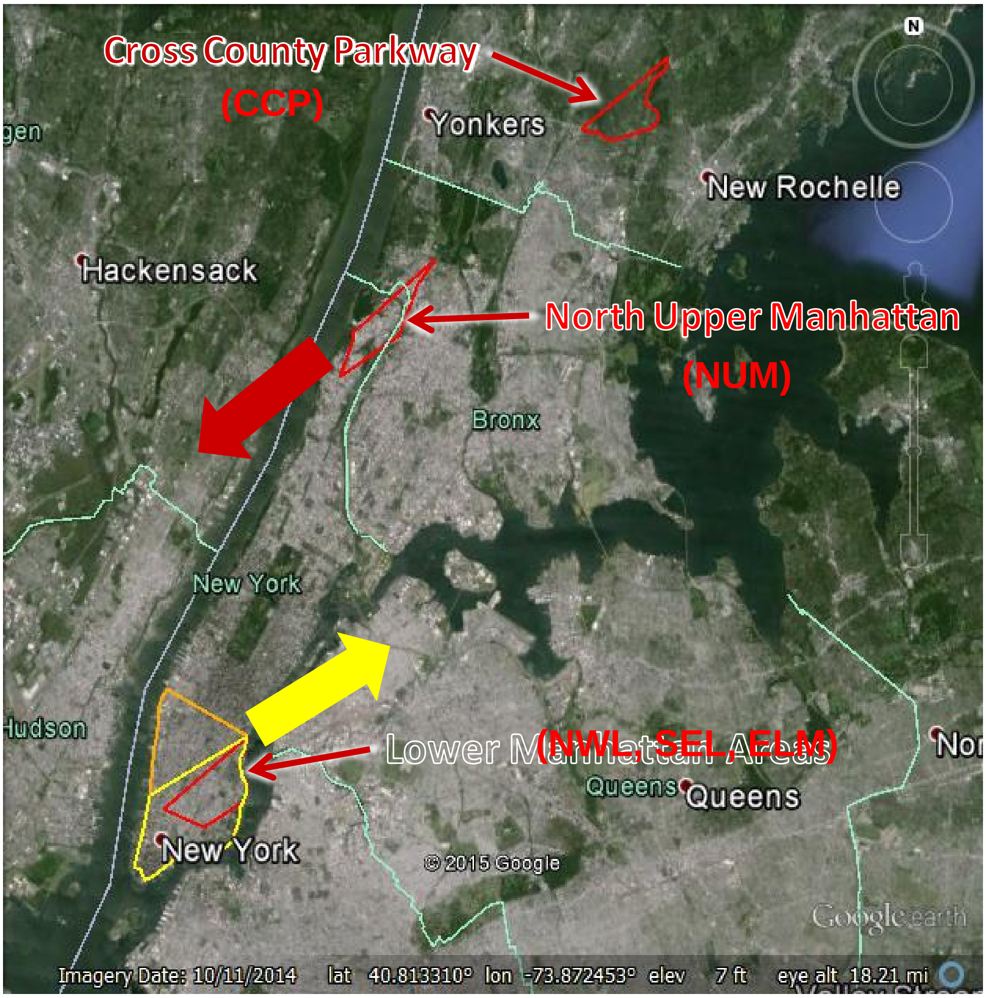

Figure 3. Areas with sufficient methane survey data to support experimental application of simple mass balance emissions estimate method. Bold yellow and red arrows indicate predominant wind direction on days for which there was available methane survey data for areas marked with the same colors (yellow or red). There were 3 areas in Lower Manhattan (see Figure 4).

Figure 4. Areas in Lower Manhattan with sufficient methane survey data to support experimental application of simple mass balance emissions estimate method. Bold yellow and red arrows indicate predominant wind direction on days for which there was available methane survey data for areas marked with the same colors (yellow or red). Bold arrows also indicate area sides where no methane data was collected, i.e., sides between the upwind and downwind sides.

Methane Emissions Estimate Areas

Cross County Parkway (CCP) in Brooklyn inbound to Manhattan, 27 November 2012: upwind run (11:26-11:28 AM, N=381, 40.94918, -73.7985 to 40.92729, -73.81230) north and downwind run (11:29:09-11:29:58 AM, N=180, 40.92323 -73.82270 to 40.92662 73.83547) west of a right angle turn in the Parkway around lat. 40.9225 long. -73.8170. Wind from 60° at 2.7 $m \cdot s^{-1}$. PBL height at 50m. Cross wind dimension 1 km. Land area 2.57 km^2. Simple difference between well mixed air 50th percentile methane concentrations (downwind – upwind) = 0.016 ppm indicated a methane emissions rate of 0.6 $\mu g \cdot m^{-2} s^{-1}$. Due to remoteness from the weather station and ceilometer in Manhattan, the PBL and wind data were considered most likely sources of error.

North Upper Manhattan (NUM), 27 November 2012, downwind run on Broadway from 181st Street (12:29-12:36, N= 936, 40.85028 -73.93581) to Thayer Street (40.86494 -73.92850), upwind reference run in Brooklyn on Major Deegan Expressway along east side of Harlem River (11:36-11:40, N= 481, 40.8860 -73.8967 to 40.8632 -73.91141). Wind from 50° at time/direction adjusted speed of 0.6 to 1.2 $m \cdot s^{-1}$. PBL height at 55m. Cross-wind dimension = 1.17km. Land area = 2.5 km^2. Simple difference between well mixed air 50th percentile methane concentrations (downwind – upwind) = 0.11 ppm indicated a methane emissions rate of 1 to 3 $\mu g \cdot m^{-2}s^{-1}$, with uncertainties again due primarily to remoteness from sources of wind and PBL data.

Northwest Lower Manhattan (NWL), 29 November 2012, downwind run the length of 34th Street (16:59-17:22, N=2751, 40.74439 -73.97461 to 40.775710 -74.00493), upwind reference run on Lincoln Highway from 34th to Canal Streets (17:22-17:36, N=1196 , 40.75706 -74.00496 to 40.72647 -74.0111). Wind from 240° at 3.6 $m \cdot s^{-1}$. PBL height at 800m. Cross-wind dimension = 2.75km. Land area = 5.2 km^2. Simple difference between well mixed air 50th percentile methane concentrations (downwind – upwind) = 0.111 ppm indicated a methane emissions rate of 110 $\mu g \cdot m^{-2}s^{-1}$.

Southeast Lower Manhattan (SEL), 29 November 2012, downwind run the length of the FDR Drive from near the East River Park Amphitheater to 34th Street (16:25-16:59, N=4980, 40.71157 -73.97845 to 40.74438 -73.97459), upwind reference run on Lincoln Highway from Canal Street southward continuing eastward onto FDR Drive to near the East River Park Amphitheater (16:17-16:25, N=1319, 40.72647 -74.0111 to 40.71153 -73.97850). Wind from 240° at 3.6 $m \cdot s^{-1}$. PBL height at 800m. Cross-wind dimension = 2.75km. Land area = 9.2 km^2. Simple difference between well mixed air 50th percentile methane concentrations (downwind – upwind) = 0.109 ppm indicated a methane emissions rate of 62 $\mu g \cdot m^{-2}s^{-1}$.

East Lower Manhattan (ELM), 30 November 2012, downwind run on Grand Street from West Broadway to Essex Street (15:44:12-15:45:42, N=2369, 40.72400 -73.97337 to 40.73491 -73.97393), upwind reference run on FDR from near 10th Street to near 23rd Street along East River (13:37-14:20, N=156, 40.72241 -73.00371 to 40.71729 -73.98892). Wind from 60° at time/direction adjusted speed of 1.3 $m \cdot s^{-1}$. PBL height at 1000m. Cross-wind dimension = 1.09km. Land area = 2.35 km^2. Simple difference between well mixed air 50th percentile methane concentrations (downwind – upwind) = 0.266 ppm indicated a methane emissions rate of 110 $\mu g \cdot m^{-2}s^{-1}$.

Table 1. Parameter Values and Methane Emissions Estimates
Areas in or near New York City, New York

Emissions Estimate Area	Wind Dir.	Wind Speed ($m\cdot s^{-1}$)	PBL Height (meters)	X-wind length (km)	Land Area (km^2)	Estimated Methane Emissions Rate ($\mu g\cdot m^{-2}s^{-1}$)
Cross County Parkway (CCP)	60	2.7	50	1	2.57	0.6
North Upper Manhattan (NUM)	50	0.6-1.2	55	1.17	2.5	1-3
East Lower Manhattan (ELM)	60	1.3	1000	1.09	2.35	110
Northwest Lower Manhattan (NWL)	240	3.6	800	2.75	5.2	110
Southeast Lower Manhattan (SEL)	240	3.6	800	2.75	9.2	62

Given the uncertainties in wind and PBL data and the coarseness of our estimates of the dimensional parameters of the emissions estimate areas, the emissions estimates were all plausible or consistent. The estimated methane emissions rates for the Cross County Parkway and North Upper Manhattan areas from 27 November data are comparable to each other and rates reported for similar urban/suburban settings by others (Jackson, 2014; Mays et al., 2009; McKain et al., 2015). The CCP area was remote from the Central Park weather station and the ORSL ceilometer location. The height of the PBL was changing rapidly during the methane survey of the North Upper Manhattan area. Consequently, emissions rates were reported as ranges because of uncertainties in the wind and PBL height data.

The 3 Lower Manhattan areas emissions estimates were in good agreement, the NWL and ELM emissions being equal at 110 $\mu g\cdot m^{-2}s^{-1}$. The estimate for the SEL area at 62 $\mu g\cdot m^{-2}s^{-1}$ was 45% lower than those for the NWL and ELM areas. This lower result may be an example of the potential error inherent in use of only well mixed air methane concentration data. Methane in poorly mixed air on the downwind side of a target area is part of the emissions within that target area. The well-mixed-air-only restriction necessarily imposes the potential for underestimating total emissions if a locally potent source is close to the downwind side of the target area. The ELM area was contained within the SEL area, and both contained the Con Ed East River Generating Station. The Generating Station was just inside the upwind side of the ELM, and just inside the downwind side of the SEL.

Methane from the Station would have been undetected on the upwind side and well mixed into the air on the downwind side of ELM and not removed by the well-mixed-air data filtration. In contrast, in the SEL the Station was just inside the downwind side, hence, poorly mixed, and recognized and removed by the well-mixed-air data filtration. Direct examination of the data indicated a substantial, typical, poorly mixed air methane excursion downwind of the Station in the SEL data set. Further, when the SEL emissions estimate was calculated without applying the well-mixed-air-only data filter the estimate increased to 125 $\mu g \cdot m^{-2} s^{-1}$, only 14% higher than the estimates for the ELM and the NWL. This result supports the likely conservative validity of the emissions estimates for Lower Manhattan (below 34th Street) at 110 $\mu g \cdot m^{-2} s^{-1}$, and suggests the East River Generating Station is a substantial local source of methane emissions.

Sources of Error

The proposed simple mass balance method requires measurement input values for wind speed, planetary boundary height, wind-run and cross-wind dimensions of the target area, and the difference between upwind and downwind well mixed air methane levels. We estimate the error in the well mixed methane measurements to be 0.5%. We estimate the relative errors for wind speed, PBL height and wind-run and cross-wind dimensions of the target area to be, respectively, 20%, 10%, 5% and 5%. Error propagation for a simple product model implies an overall error in our emissions estimate of less than 25%. The calculation of the whole island estimated emissions is a further simple product, which, assuming a 30% error in our estimated distribution of emissions over the island, leads to a propagated error of less than 40%. Further development of the method and supporting data collection efforts can be expected to substantially reduce these errors.

Plausibility of Results -- Comparison to Other Methods in Other Cities

The accuracy of the method is also potentially affected by three other factors: (i) the exclusion of poorly mixed air methane data, (ii) the assumption of effectively uniform vertical mixing throughout the PBL and (iii) that no methane is transferred through the top of the PBL. Exclusion of poorly mixed air methane data (i) is likely to cause considerable underestimate of emissions. This was illustrated by the results for the SEL and the ELM areas as discussed above. Exclusion of poorly mixed air data caused emissions in the SEL to be underestimated by an apparent 44%, while inclusion caused an apparent 13% overestimate, compared to the ELM. (ii) If the methane were transferred upward through the top of the PBL the assumption of no transfer would also cause an underestimate of emissions. (iii) The limited data and information we have suggests that vertical mixing in the PBL is probably complete over Manhattan. If it were not, then the emissions estimate would be overestimated, but not likely by more than 25%. Hence, the emissions estimates we calculated for Manhattan are more likely to be too low than too high. The impacts of these factors can be managed in further development of the method.

We sought to evaluate the plausibility of our emissions estimates for Lower Manhattan. We were unable to locate other methane emissions estimates based on actual methane measurements for Lower Manhattan or other sub-city urban areas. As an alternative we undertook a comparison of our Manhattan results to those reported for other cities. To make such a comparison it was necessary to get all the cities methane emission rates onto the same basis.

We extrapolated the Lower Manhattan emissions rates to generate an emission rate for all of Manhattan by applying the relationship developed from our general analyses of methane survey data for all of Manhattan, i.e., that half of all emissions on the island occur south of 42nd Street. The extrapolated whole-island Manhattan methane emission rate was 66 $\mu g \cdot m^{-2} s^{-1}$, which is the equivalent of 6.6 billion cubic feet of methane per year, or about 6.6% of the volume of natural gas used in Manhattan per year.

We located reports of related methane emissions estimates for 5 other cities (Table 2), Krakow, Poland (Kuc, Rozanski, Zimnoch, Necki & Korus, 2003; Zimnoch et al., 2010), London, UK (Helfter, Nimitz, Barlow & Wood, 2013), and in the U.S.A., Boston, Massachusetts (McKain et al., 2015), Indianapolis, Indiana (Mays et al., 2009), and an unidentified 'Small Town' in the US Midwest (Lamb et al., 1995). Zimnoch et al. (2010) measured PBL height by SODAR (sonic distance and ranging) and methane using gas chromatography and selected nighttime data to take advantage of stable PBL conditions. Helfter et al. (2013) used eddy covariance carbon dioxide and methane data and methane/carbon dioxide ratios to back calculate over time methane emissions for central London. Lamb et al. (1995) used a tracer release technique to estimate methane emissions. McKain et al used 4 static CRDS installations to measure methane levels for a year at 2 locations in Boston, 1 reference site outside Boston and 1 reference site outside the Boston urban region. Mays et al. (2009) used an aircraft-borne CRDS and other instrumentation to assess methane concentration, wind speed, PBL height, etc. The reported methane emissions rates for those four cities ranged from 0.43 to

2 $\mu g \cdot m^{-2} s^{-1}$, considerably lower than our estimate for Manhattan at 66 $\mu g \cdot m^{-2} s^{-1}$. However, the five cities range widely in population density (66,000 persons km^{-2} in Manhattan to 440 in the Boston urban region) and gross annual NG consumption (12330 Gg yr^{-1} in the Boston urban region to 49 Gg yr^{-1} in 'Small Town').

In urban areas, the intensity of NG usage will be related to population. Gas flow rates and size and complexity of gas distribution systems can be reasonably expected to increase in proportion to the number of customers being served. With a daytime population density of around 66,000 persons km^{-2}, Manhattan is 6 times more densely populated than the next densest, London, at 11,000 persons km^{-2} (Helfter et al., 2013; Moss & Qing, 2012). Those populations are served with NG, but delivery and use efficiencies can be expected to vary among cities. As a more concise approach we examined total NG usage per unit area for the 5 cities, and compared them on an equivalent gas usage density basis (Table 2).

Table 2. Natural Gas Usage and Methane Emissions in Five Cities.

City	**Natural Gas Usage**		**Methane Emissions**		
	Annual	Area•time density basis	Area•time density basis	On a Manhattan gas usage density basis	As percent of usage density
	$Gg\ yr^{-1}$	$\mu g\ m^{-2}\ s^{-1}$	$\mu g\ m^{-2}\ s^{-1}$	(Units[a])	%
Manhattan NYC US 2012	1800	1000	66	66	6.6
Indianapolis IN US 2008	920	28	2.2	79	7.9
"Small Town" US	49	68	2.2	32	3.2
Boston Urban Region US ’12-‘13	12330	22	0.59	27	2.7
Krakow Poland '96-'97	220	22	0.62	28	2.8
Krakow Poland '05-'08	300	29	0.43	14	1.4
London (4 boroughs) UK 2012	610	260	2.2	8	0.8

[a]µg methane emissions $m^{-2}s^{-1}$ per 1000 µg NG usage $m^{-2}s^{-1}$

The five cities range widely in gross annual NG usage (Gg yr-1). When NG usage is considered on an area-time density basis ($\mu g{\cdot}m^{-2}s^{-1}$), Manhattan usage density (1000 $\mu g{\cdot}m^{-2}s^{-1}$), is nearly 4 times denser than that of London ($\mu g{\cdot}m^{-2}s^{-1}$), almost 36 times denser than that of Indianapolis (28 $\mu g{\cdot}m^{-2}s^{-1}$), more than 45 times denser than that of the Boston urban region. The Indianapolis emissions/usage-density ratio 79 was the highest. The ratio for Manhattan was 66. “Small town”, Krakow in 1996-1997 and the Boston urban region had similar ratios of 32, 28, and 27, respectively. Krakow in 2005-2008 and London had substantially lower ratios at 14 and 8, respectively. Zimnoch et al. (2010) suggested the apparent decrease in emissions in Krakow between 1996-1997 and 2005-2008 may have been due to a gas infrastructure improvement program. Given the age, size and complexity of the NG gas distribution system in Manhattan, both the emissions/usage-density ratio and, hence, our emissions estimate of 66 $\mu g{\cdot}m^{-2}s^{-1}$ appear plausible.

Implications

Methane emissions from NG infrastructure have become a matter of some concern, especially with respect to the proposition that increased use of NG in place of other fossil fuels will result in lower GHG emissions per unit of useful energy. Investigations of urban methane emissions have consistently found much of the methane is from the local NG system (Zimnoch et al., 2010; Jackson et al., 2014; Belluci et al., 2012). Other recognized potential methane sources include automobiles and other combustion sources, sewage systems, landfills, wetlands, agricultural facilities. There are no agricultural facilities, wetlands, sewage treatment plants or waste landfills in Lower Manhattan. Using the automobile methane emissions rate (1.5 x 10^{-4} g CH4 per g CO2) of Nam, Jensen and Wallington (2004) and commuter and traffic estimates (Moss & Qing, 2012) we estimated vehicle methane emissions would not likely exceed 2 $\mu g{\cdot}m^{-2}s^{-1}$ in Manhattan, and the total methane emissions due to incomplete combustion in vehicle and non-vehicle sources should not exceed 5 $\mu g{\cdot}m^{-2}s^{-1}$. Consequently, we concluded it is likely that something approaching 90% of our estimated

66 $\mu g{\cdot}m^{-2}s^{-1}$ methane emitted in Manhattan was likely from the NG system, which will include leaks at point of use.

It appears that in most cities with NG service, most of the methane emissions are the collective effect of local NG infrastructure and utilization technology, and, consequently represent the distribution system component of total leakage for the NG production-transmission-distribution system. Considering methane emissions (NG leakage) on a NG-usage-area-density basis (Table 2) eliminates the need to consider all possible types of NG leaks (compressors, regulators, valves, lines, point of use, etc.). Methane emissions in the 5 cities ranged from 0.8% to 7.9% of the gas usage. In Indianapolis and Manhattan methane emissions amounted to 6.6%-7.9% of gas usage. Even in the Boston urban region, which includes large non-urban areas, methane emissions were 2.7% of the gas used.

The greenhouse gas (GHG) impact break-even point for NG-versus-coal (for power production) has been estimated at a total NG system leakage rate of 2.8% of total NG production from well drilling through delivery and use (Alvarez et al., 2013; Howarth,2014). Based on methane emissions estimates for the 5 globally prominent cities and a small US town, 2/3 of urban NG distribution systems exceed the GHG (electric power production) break-even gas leakage for the entire NG production-through-consumption system. None of the 4 US cities had a leakage rate below the 2.7%. Even for the US city with the lowest likely NG-related methane emission rate there is no plausible leakage rate for the upstream segments (production and transmission) of that NG stream that would allow NG to be construed as GHG advantageous with respect to other fossil fuels for electric power production. The dominant use of natural gas in the US (>60%) is combustion to generate heat for residential and industrial purposes, not electric power production. The energy conversion efficiency advantages of gas for electric power production do not apply to combustion for heat production. Consequently, the methane emissions (NG leakage) rates that support a GHG advantage for natural gas over other fossil fuels are much lower, making use of NG for heating GHG-disadvantageous at the emissions rates reported for the 6 cities we considered.

Conclusion

We developed a generally applicable, simple method for calculating methane emissions from distributed ground level ambient air methane concentration, weather and PBL data. Methane data for Manhattan showed concentrations consistently increased from upwind to downwind areas on the island. The method provided plausible consistent estimates of methane emissions in Lower Manhattan even though methane survey data were collected under different wind conditions and in nested or separate neighboring areas. Our simple calculation based on changes in methane concentrations, wind speed, height of the planetary boundary layer and observed relative differences between the northern and southern parts of the island generated an estimated methane emissions rate of 66 $\mu g{\cdot}m^{-2}s^{-1}$ for the whole island, and plausible rates for 2 less densely populated areas in the region. We examined methane emissions as a function of NG usage density to compare the Manhattan emissions rate indicated by our method to rates reported for 5 other cities by other investigators using other methods. Our estimated emission rate for Manhattan was within the range of rates among the other 5 cities. The emissions estimates for the 6 cities indicated use of natural gas in lieu of other fossil fuels will not provide any climate change advantage, and will likely do more harm than good. We conclude the proposed simple mass balance methane emissions estimation method based on mobile CRDS data produced plausible results even with only opportunistic data sets. The method should be considerably more effective if wind, PBL, and methane data can be collected in data surveys designed for the purpose of estimating emissions. Further efforts to apply the method opportunistically with respect to other previously collected data sets could rapidly generate measurement-based methane emissions estimates for numerous other areas. More fully developed and applied the method could substantially contribute to meeting the need for methane emissions data, and should be adaptable to other trace gases for which similar mobile measurement capabilities are available.

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References:

Alvarez, R. A.; Pacala, S. W.; Windbrake, J. J.; Chameides, W. L.; Hamburg, S. P., Greater focus needed on methane leakage from natural gas infrastructure. *Proceedings of the National Academy of Sciences of the United States of America* **2014,** *109* (17), 6435-6440.

Bellucci, F.; Bogner, J. E.; Sturchio, N. C., Greenhouse Gas Emissions at the Urban Scale. *Elements* **2012,** *8*, 445-449.

Cade, B. S., & Noon, B. R. (2003). A gentle introduction to quantile regression for ecologists. *Frontiers in Ecology and the Environment*, *1*(8), 412-420.

Dlugokencky, E. J.; Nisbet, E. G.; Fisher, R.; Lowry, D., Global atmospheric methane: budget, changes and dangers. *Philos Trans A Math Phys Eng Sci 369* (1943), 2058-72.

EPA-OIG (2013). U.S. Environmental Protection Agency-- Office of Inspector General. *EPA Needs to Improve Air Emissions Data for the Oil and Natural Gas Production Sector*. Report No. 13-P-0161. February 20, 2013.

Fowler, D., Experimental designs appropriate for flux determination in terrestrial and aquatic ecosystems In *Developments in Atmospheric Science -- Approaches to Scaling of Trace Gas Fluxes in Ecosystems*, Bouwman, A. F., Ed. Elsevier BV: Amsterdam, 1999; Vol. 24.

Gan, C.-M.; Wu, Y.; Gross, B. M.; Arend, M.; Moshary, F.; Ahmed, S., A comparison of estimated mixing height by multiple remote sensing

instruments and its influence on air quality in urban regions. In *IEEE International Geoscience & Remote Sensing Symposium, IGARSS 2010*, Honolulu, Hawaii, USA, 2010.
Helfter, C.; Nemitz, E.; Barlow, J. F.; Wood, C. R., Carbon dioxide and methane emission dynamics in central London (UK). *Geophysical Research Abstracts* **2013,** *15* (EGU2013), 7036.
Howarth, R.W. (2014) A bridge to nowhere: methane emissions and the greenhouse gas footprint of natural gas. *Energy Science and Engineering.* (open access, accepted 22 April 2014). doi: 10.1002/ese3.35.
Jackson, R. B.; Down, A.; Phillips, N. G.; Ackley, R. C.; Cook, C. W.; Plata, D. L.; Zhao, K. 2014. Natural Gas Pipeline Leaks Across Washington, DC. *Environ Sci Technol 48* (3), 2051-8. doi: 10.1021/es404474x.
Koenker, R., & Hallock, K. (2001). Quantile regression: An introduction. *Journal of Economic Perspectives*, *15*(4), 43-56.
Kuc, T.; Rozanski, K.; Zimnoch, M.; Necki, J. M.; Korus, A., Anthropogenic emissions of CO2 and CH4 in an urban environment. *Applied Energy* **2003,** *73*, 193-203.
Lamb, B. K.; McManus, J. B.; Shorter, J. H.; Kolb, C. E.; Mosher, B.; Harriss, R. C.; Allwine, E.; Blaha, D.; Howard, T.; Guenther, A.; Lott, R. A.; Siverson, R.; Westburg, H.; Zimmerman, P., Development of atmospheric tracer methods to measure methane emissions from natural gas facilities and urban areas. *Environ Sci Technol* **1995,** *29* (6), 1468-79.
Legbandt, T. NYCMetNet. http://nycmetnet.ccny.cuny.edu.
Mays, K. L.; Shepson, P. B.; Stirm, B. H.; Karion, A.; Sweeney, C.; Gurney, K. R., Aircraft-based measurements of the carbon footprint of Indianapolis. *Environ Sci Technol* **2009,** *43* (20), 7816-23.
McKain, K., Down, A., Raciti, S.M, Budney, J., Hutyra, L.R., Floerchinger, C., Herndon, S.C., Nehrkorn, T., Zahniser, M.S., Jackson, R.B., Phillips, N., Wofsy, S.C. 2015. Methane emissions from natural gas infrastructure and use in the urban region of Boston, Massachusetts. *Proceedings of the National Academy of Sciences of the United States of America* 112(7) 1941–1946. doi: 10.1073/pnas.1416261112.
Moss, M. L.; Qing, C. *The Dynamic Population of Manhattan*; Rudin Center for Transportation Policy and Management, Wagner School of Public Service, New York University: New York, New York, 2012.
Nam, E. K.; Jensen, T. F.; Wallington, T. J., Methane Emissions from Vehicles. *Environmental Science and Technology* **2004,** *38*, 2005-2010.
Payne Jr., B.F. & Ackley, R. (2012, December 16). Report on a Preliminary Investigation of Ground-Level Ambient Methane Levels in Manhattan, New York City, New York. Retrieved from http://documents.dps.ny.gov/public/Common/ViewDoc.aspx?DocRefId={BD0C41AF-925B-425E-A17A-1EF5ABAFAC19}.

Payne Jr., B.F. & Ackley R. (2013, March 11). Extended Report on a Preliminary Investigation of Ground-Level Ambient Methane Levels in Manhattan, New York City, New York. Retrieved from http://www.damascuscitizensforsustainability.org/wp-content/uploads/2013/03/Manhattan-Final-report.pdf.

Phillips, N. G.; Ackley, R.; Crosson, E. R.; Down, A.; Hutyra, L. R.; Brondfield, M.; Karr, J. D.; Zhao, K.; Jackson, R. B. 2012. Mapping urban pipeline leaks: methane leaks across Boston. *Environ Pollut 173*, 1-4. doi: 10.1016/j.envpol.2012.11.003.

Piironen, Antti. 1996. Atmospheric Boundary Layer Structure. Retrieved from http://lidar.ssec.wisc.edu/papers/akp_thes/node6.htm.

Stocker, T. F.; Qin, D.; Plattner, G.-K.; Tignor, M.; Allen, S. K.; Boschung, J.; Nauels, A.; Xia, Y.; Bex, V.; Midgley, P. M.; (eds.) *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*; Intergovernmental Panel on Climate Change: Cambridge, UK and New York, NY, USA, 2013.

The White House (2014). *Climate Action Plan: Strategy to Reduce Methane Emissions.* March 2014.

Zimnoch, M.; Godlowska, J.; Necki, J. M.; Rozanski, K., Assessing surface fluxes of CO2 and CH4 in urban environment: a reconnaissance sudy in Krakow, Southern Poland. *Tellus* **2010,** *62B*, 573-580.